Density Functional Theory of Multicomponent Quantum Dots

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Abstract

Quantum dots with conduction electrons or holes originating from several bands are considered. We assume the particles are confined in a harmonic potential and assume the electrons (or holes) belonging to different bands to be different types of fermions with isotropic effective masses. The density functional method with the local density approximation is used. The increased number of internal (Kohn-Sham) states leads to a generalisation of Hund's first rule at high densities. At low densitites the formation of Wigner molecules is favored by the increased internal freedom.

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I. INTRODUCTION

In simple models of quantum dots, the conduction electrons (or holes) of a semiconductor are confined into a two-dimensional harmonic trap (for reviews see [1, 2]). The band structure of the material is taken into account through the effective mass approximation, and screening effects are accounted for by the dielectric constant. The problem is then reduced to solving the many-particle problem of interacting electrons in a two-dimensional harmonic potential. The electrons have spin as an 'internal degree of freedom'. Neglecting spin-orbit coupling, the spin-up and spin-down electrons can be treated as separate interacting particles, and consequently we can say that the normal electron gas is a two-component gas, the components being the spin-up and spin-down electrons. Similarly, we will call the polarised electron gas as a one-component system (sometimes also called a system of spin-less fermions).

The simple picture will fail in describing more complex structures where the number of degrees of freedom of the electrons is increased either by several two-dimensional layers or by multiple valleys of the band structure. For example, in a vertical double-layer quantum dot the electrons confined in the two layers form (in the vertical direction) "odd" and "even" states [3, 4, 5] which could be approximated as different components, or as different isospin-states of the electron [6]. The isospin together with the spin would make the system a four-component electron gas.

Other multicomponent electron systems would be quantum dots in silicon. In this case the conduction electrons originating from four valleys of the conduction band could be approximated as different, but mutually interacting fermions. Similarly, quantum dots with holes would always have particles belonging to different bands, i.e. heavy holes and light holes [7].

Density functional theory in the local (spin) density approximation (LSDA) provides a flexible method to study the ground state properties of interacting electrons in quantum dots [2]. In LSDA the exchange and correlation effects of the interacting conduction electrons are locally approximated by the exchange-correlation energy (ϵ_{xc}) of the two-dimensional, homogenous gas. Similarly, in a multicomponent electron system the starting point for the local density approximation is the exchange-correlation energy of a two-dimensional multicomponent gas. Recently, we have suggested that the multicomponent ϵ_{xc} can well be

approximated by extending the parametrised two-component function of Attaccalite *et al.* [8] to a higher number of components [9]. This parametrisation was shown to have correct high and low density behaviour and it has rather simple dependence on the densities of different components. Moreover, the dependence of the effective masses of the components can be approximated through a simple scaling of the density parameter by an effective average mass.

In this paper we study the electronic structure of quantum dots where the electron gas has from one up to eight internal degrees of freedom, using the exchange-correlation functional suggested in Ref. [9]. The inclusion of more components increases the number of internal degrees of freedom in the system. As a result, in the self-consistent scheme the electrons can access more Kohn-Sham states in addition to the usual two spin states, leading to new features in the electronic structure. We will discuss the electronic shell structure of four-component quantum dots by studying the addition energy spectrum at high densities. In the low density limit the formation of Wigner molecules in an eight-component quantum dot is investigated. We found that the seven-electron configuration is particularly stable like the classical point-charge calculation predicts [12]. In the Wigner molecule limit the fermions of different components are distributed spatially so that antiferromagnetic frustration is avoided. Finally we notice that even a slight increase of mass favors the heavier components as the heavy component states are pushed down in energy.

II. THEORETICAL MODEL

We write the total density of the multicomponent electron gas as

$$n(\mathbf{r}) = \sum_{i=1}^{\Lambda} n_i(\mathbf{r}) = n(\mathbf{r}) \sum_{i=1}^{\Lambda} \nu_i(\mathbf{r}), \tag{1}$$

where Λ is the number of components, n_i are the densities of components and ν_i dimensionless concentrations of the components. In the multicomponent case, the use of concentrations is simpler than the use of total density and polarization ζ, as usually done for the normal electron gas (in the two-component case, $n_1 = n_\uparrow$ and $n_2 = n_\downarrow$ and $\zeta = \nu_1 - \nu_2$). Following the notations of Ref. [9] we define numbers

$$Z_\gamma = \sum_{i=1}^{\Lambda} \nu_i^\gamma, \tag{2}$$

which allows us to write the exchange-correlation functional of Attaccalite *et al.* [8] as

$$\epsilon_{xc}(r_s, \{\nu_i\}) = e^{-\beta r_s}[\epsilon_x - \epsilon_x^{(6)}] + \epsilon_x^{(6)} + \alpha_0(r_s) + \alpha_1(r_s)(2Z_2 - 1) + \alpha_2(r_s)(2Z_2 - 1)^2 \ , \quad (3)$$

where $\epsilon_x^{(6)} = (1 + \frac{3}{8}(2Z_2 - 1) + \frac{3}{128}(2Z_2 - 1)^2)\epsilon_x(r_s, \zeta = 0)$. As shown earlier [9] this analytic continuation of the originally two-component functional approximates very well all existing results for the exchange-correlation energy of a multicomponent electron gas.

The exchange energy is independent of the particle mass. If the effective masses of all the particles are the same, the mass dependence of the total energy becomes just a scaling factor. If the masses are not the same we can use as a first approximation a properly weighed average mass as a scaling factor and write [9]

$$\epsilon_{xc}(r_s, \{\nu_i\}, \{m_i\}) = \frac{M}{m_e}\epsilon_{xc}(Mr_s, \{\nu_i\}, \{m_i = m_e\}) \ , \quad (4)$$

where m_e is the bare mass of the electron (or any suitably chosen effective mass), m_i is the effective mass of component i, and M is an average mass defined as

$$\frac{1}{M} = \frac{1}{Z_2}\sum_i^{\Lambda} \frac{\nu_i^2}{m_i}. \quad (5)$$

Note that the density parameter r_s in our formulation always refers to the total number density of all particles: $r_s = 1/\sqrt{\pi n}$.

The Kohn-Sham equations have to be solved self-consistently for each component of the electron gas. The effective potential $V_{\mathrm{eff},i}$ consists of the external harmonic confinement (assumed here to be the same for all components), of the Coulomb repulsion of the electron density distribution (Hartree term) and of the exchange-correlation potential, which can be directly derived from the multicomponent exchange-correlation energy:

$$V_{\mathrm{eff},i}(\mathbf{r}) = \frac{1}{2}Kr^2 + \int d\mathbf{r}' \frac{e^2 n(\mathbf{r}')}{4\pi\varepsilon_0\epsilon|\mathbf{r} - \mathbf{r}'|} + V_{\mathrm{exc},i}(r_s(\mathbf{r}), \{\nu_i(\mathbf{r})\}, \{m_i\}), \quad (6)$$

where K is the strength of the external confinement, ϵ the dielectric constant. Note that the exchange-correlation potential depends locally on the concentrations of each component. The Kohn-Sham equations to be solved simultaneously for all components are

$$-\frac{\hbar}{2m_i}\nabla^2\psi_{i,k}(\mathbf{r}) + V_{\mathrm{eff},i}(\mathbf{r})\psi_{i,k}(\mathbf{r}) = \epsilon_{i,k}\psi_{i,k}(\mathbf{r}), \quad (7)$$

resulting in the densities n_i of all components i,

$$n_i(\mathbf{r}) = \sum_k^{N_i} |\psi_{i,k}(\mathbf{r})|^2 \, , \quad (8)$$

where N_i is the number of electrons of component i. In the ground state the lowest single particle levels are filled and the numbers N_i are known only after the ground state is found. In practice we solve the equations by keeping the numbers N_i fixed and then choose the configuration which gives lowest total energy.

The Kohn-Sham equations were solved using a plane-wave expansion and the fast Fourier transform technique. We use up to 23×23 plane waves and, correspondingly a 45×45 lattice at which the density and potential is derived. When iterating the Kohn-Sham equations, a mixing of the new and old potential is necessary to obtain convergence. The convergence is slow especially at low densities where broken symmetry solutions (localization of electrons to Wigner molecules) emerge. We use effective atomic units where energy is given in effective Hartree, Ha*=$m_e^* e^4/\hbar^3(4\pi\epsilon_0\epsilon)^2$ and the unit of distance is the effective Bohr radius $a_0^* = \hbar^2 4\pi\epsilon_0\epsilon/m_e^* e^2$.

III. RESULTS

A. Shell structure – Addition spectrum

As confined, fermionic quantum systems, quantum dots show shell structure with degeneracies determined by the symmetries of the confining potential. The independent electron energy spectrum for a 2D harmonic oscillator is $\varepsilon_{nl} = \hbar\omega_0(2n + |l| + 1)$, where n is the principal quantum number and l is the angular momentum (or its z-projection). At high densities the independent electron scheme describes the qualitative characteristics of the shell filling of the interacting system reasonably well, as known from the earlier studies [2] of normal quantum dots. The main effect of the electron-electron interaction is to produce the spin determined by Hund's first rule [11]. The shell structure effects are usually shown in a form of addition energy spectrum where the second difference of the total energy is plotted as a function of the number of electrons in the dot. Addition energies measure the changes $\mu(N+1) - \mu(N)$ in the chemical potential $\mu(N) = E(N) - E(N-1)$.

We will first study the addition energy spectrum of an ideal four-component electron system in a quite high electron density, $r_s = 2\ a_0^*$. A physical realization of such system could be a vertical double dot, where the interlayer distance is very small. Figure 1 shows the addition energy spectrum and a schematic picture of the single particle levels. Since

each single particle level can now occupy four electrons the closed shells correspond to total electron numbers 4, 12, 24, etc. These 'magic' numbers are seen as pronounced maxima in the addition energy spectrum. In between, we see smaller maxima at every even electron number up to 12 electrons and at every third electron number between 12 and 24. These are manifestations of Hund's first rule generalized to the multicomponent case: Degenerate states are filled one component at a time. This minimizes the total energy, since the exchange energy favors a 'polarized' electron gas. For example, in the third shell we see maxima at $N = 15$, 18, and 21, corresponding to filling the three orbitals of the first, second and third component of the four-component electron gas.

B. Formation of Wigner molecules

At low enough densities the electrons in the parabolic quantum dot are expected to form Wigner molecules as the correlation effects start to dominate the electronic structure. In this limit the electrons localise in the classical configuration that minimises the electrostatic repulsion [12] and this charge distribution symmetry is reflected in the internal structure of the many-body wavefunction [13]. In the multicomponent systems the localisation is eased up due to the fact that electrons can access more than two internal states. In the low density limit we study an eight component system in fixed external confinement with $K = 2 \cdot 10^{-4}$ effective atomic units. This choise corresponds to densities that are only slightly higher than those where a polarised (one-component) state becomes the ground state for different electron numbers. At these low densities the multicomponent local density approximation localises the electrons in the classically predicted configurations. Figure 2 shows the total electron densities for seven, eight and nine electrons. In the cases of seven and eight electrons, one electron is in the center and the rest form a ring around it, while in the case of nine electrons two electrons form a 'dimer' at the center and seven electrons from a ring around them. The geometries are in perfect agreement with those of classical electrons [12].

Figure 3 shows the addition energy spectrum of the eight-component quantum dot at the low density. The spectrum does not any more show features of shell structure and Hund's rule, but shows a small kink at $N = 7$, in agreement with the maximum in the addition energy spectrum of the purely classical system, determined from the results of Ref. [12], and also shown in Fig. 3. Classically, the seven electrons can form a perfect hexagon with

one electron at the center. The quantum mechanical solution is the same as shown in Fig. 2. In the density functional theory the localized electrons are not point charges as in the classical case. As a consequence, the spectrum is smoother than that of the classical result.

The evolution of the ground state as a function of density parameter r_s is investigated in an eight electron quantum dot with four components. As discussed earlier, at high densities $r_s \lesssim 4\ a_0^*$ the ground state obeys Hund's rule giving the configuration (3,3,1,1) for the ground state. As the density is lowered, the electron structure shows a Wigner molecule-like state already at $r_s = 6.0\ a_0^*$. Six localised electrons are at the outer circumference with two non-localised in the middle, as shown in Fig. 4. The six electrons at the outer radius belong to two components and the two electrons in the middle occupy also two components. The electrons in the outer ring are distributed spatially so that the two nearest neigbours of each electron belong to other component. This means that the densities of the outer components are rotated by π with respect to one another. In this way the system will avoid antiferromagnetic frustration. For $r_s \gtrsim 8\ a_0^*$ the electrons localise into a classically predicted configuration with seven electrons at the outer radius and one in the middle. The frustration is again avoided by taking the nearest neighbours for each electron from other components, as shown in Fig. 4.

C. Mass dependence

The effect of the varying mass was tested in the four-component system with $r_s = 2.0\ a_0^*$ and $N = 24$. The masses of two components were increased ($m_1 = m_2 = m$) while the other two masses were kept constant ($m_3 = m_4 = 1.0\ m_e$). For $m = 1.0\ m_e$ the sd-shell is filled giving the "magic" configuration (6,6,6,6). The mass increase shifts the orbitals of the lighter components up in energy relative to heavier components, as shown in Fig.5. As a consequence, at $m = 1.2\ m_e$ the sd-orbitals of the light components are empty and the fp-orbitals of the heavier components are occupied according to Hund's rule leading to configuration (10,8,3,3). Already at $m = 1.8\ m_e$ the lighter orbitals have only two electrons and the heavier components obey Hund's rule resulting in the configuration (12,10,1,1).

IV. SUMMARY AND DISCUSSION

We have studied the general features of quantum dots confining a multicomponent electron gas. At high densities the exchange energy favors polarization of electrons and the degenerate energy levels are filled with one component at a time. This leads to an addition energy spectrum which besides the peaks at full shells also shows peaks coming from the generalization of Hund's rule.

The increased number of internal degrees of freedom of the electrons make it easier for the electrons to localize to Wigner molecules. We have demonstrated this for four- and eight-component systems. In the low density limit the addition energy spectrum then does not any more show the electronic shell structure but the geometrical shell structure of Wigner molecules.

If the different electrons (or holes) have different masses, the localization pushes the energy states of the heavy particles down as compared to those of the light particles. If the mass difference is small the addition energy spectrum is expected to be complicated due to the mixture of light and heavy particle states. However, if the mass ratio is at a typical value of heavy and light holes, say five, the light holes do not play any role until the dot has several tens of particles.

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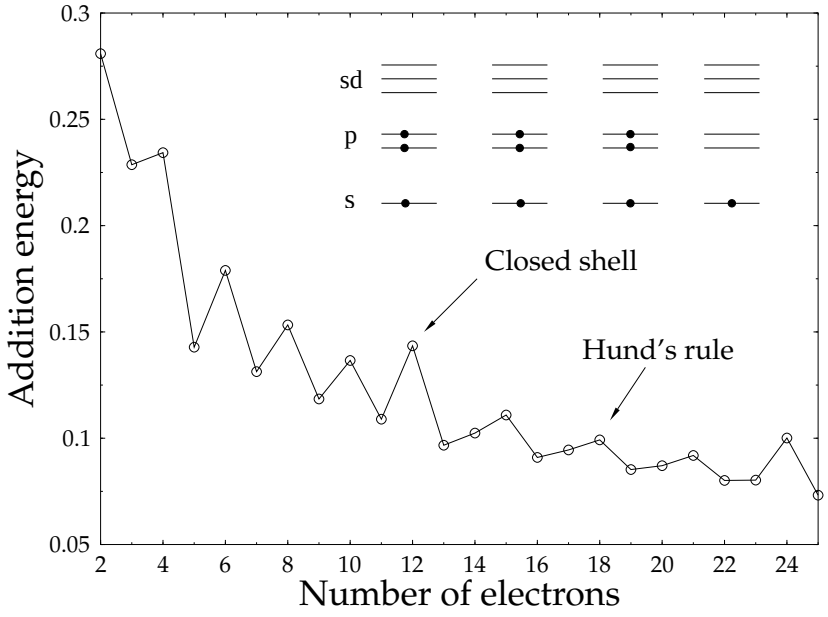

FIG. 1: Addition energy spectrum of a four-component quantum dot. The confinement potential changes with the number of electrons so that the average electron density in the dot center corresponds to $r_s = 2\ a_0^*$. The inset shows schematically the filling of levels in the case of 10 electrons. The peaks at 4, 12 and 24 are caused by shell closings while those in between are caused by Hund's rule

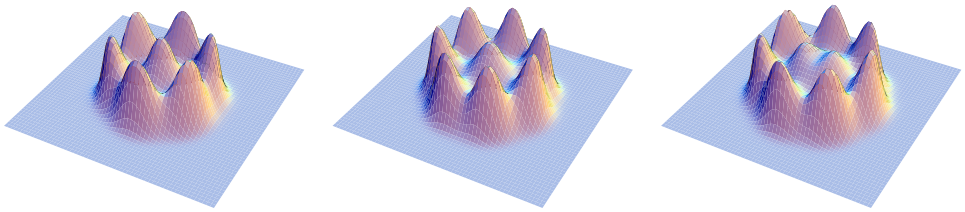

FIG. 2: The total electron densities of a quantum dot having (from left to right) 7, 8 and 9 electrons at low densities. The confinement strenght is $K = 2 \cdot 10^{-4}$ atomic units. The localization in the local density approximation is made possible by the eight internal degrees of freedom of the system.

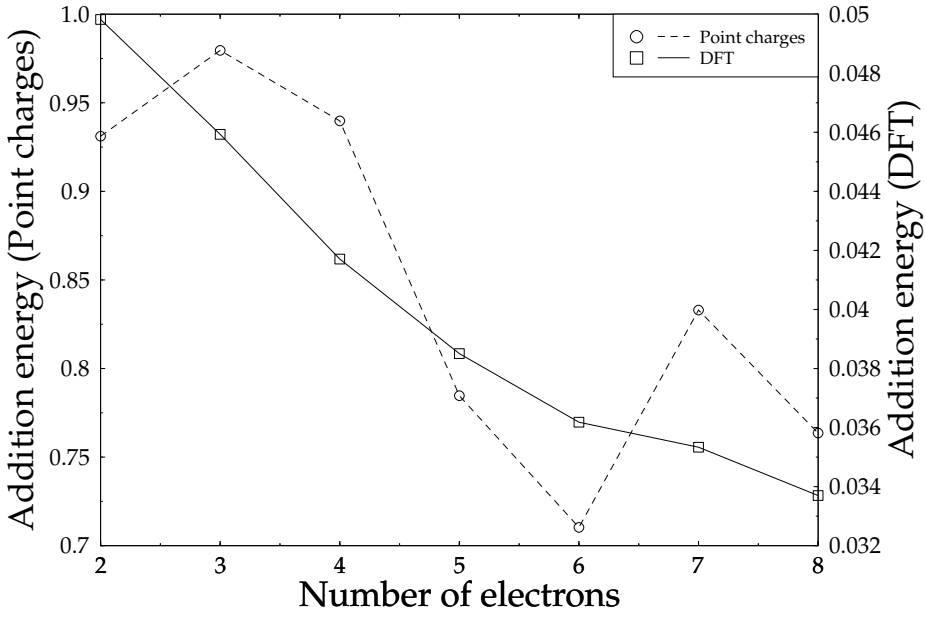

FIG. 3: Addition energy for a eight component quantum dot at a low electron density (the confinement strenght is $K = 2 \cdot 10^{-}4$ atomic units). The spectrum shows a weak kink at $N = 7$ as a precursor of the geometrically magic structure. For comparison, addition energy spectrum of classical electrons is shown.

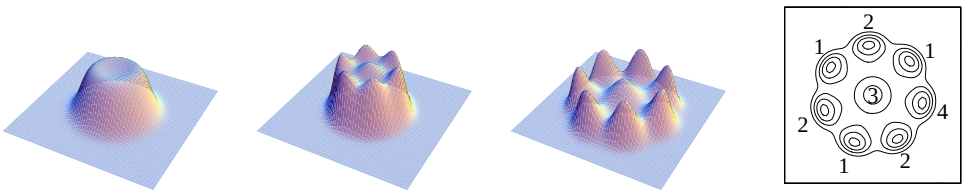

FIG. 4: Electron density of a four-cmponent quantum dot for three different values of r_s: From left to right $r_s = 2\ a_0^*$, 6 a_0^*, and 14 a_0^*. The localization in the multicomponent LDA is made possible by the fact that the neighbouring localized electrons belong to different components as indicated by numbers in the contour plot.

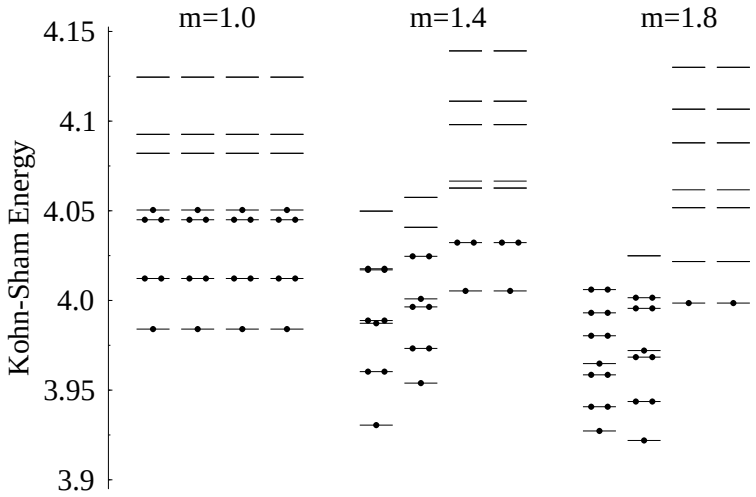

FIG. 5: The effect of varying effective mass on the single particle Kohn-Sham levels in a four component electron system with 24 electrons. The light components have mass $m = 1\ m_e$ and the heavy mass is indicated in the figure.